

# Possible Existence of Superconductivity in the Quasi-One-Dimensional Conductor $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ at Ultrahigh Magnetic Fields, $H \geq 45 \text{ T}$ .

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We derive a so-called gap equation for superconductivity in a quasi-one-dimensional (Q1D) layered conductor in a magnetic field, which is parallel to both its conducting plane and its conducting axis. This equation demonstrates that the orbital destructive effects against superconductivity cannot destroy it, if the perpendicular to the plane coherence length is less than the inter-plane distance. On the basis of our results, we suggest arguments that some triplet superconducting phase was indeed discovered in ultrahigh magnetic fields in the Q1D superconductor  $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$  [see Xiaofeng Xu et al., *Phys. Rev. Lett.*, **102**, 206602 (2009).] We also discuss a possibility that the above-mentioned superconducting phase can be stable at arbitrary high magnetic fields.

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Historically, in quasi-one-dimensional (Q1D) compounds, superconductivity was first discovered in the organic conductors with chemical family  $(\text{TMTSF})_2\text{X}$ , where  $\text{X}=\text{PF}_6$ ,  $\text{ClO}_4$ , etc. [1,2]. Note that, from the beginning,  $(\text{TMTSF})_2\text{X}$  superconductors were considered as good candidates for triplet superconducting pairing [3-7]. Nevertheless, more recently it has been found that the superconductor  $(\text{TMTSF})_2\text{ClO}_4$  is actually a singlet  $d$ -wave like one [8-10], whereas the superconductor  $(\text{TMTSF})_2\text{PF}_6$  can still be considerate as a candidate for triplet superconductivity [5-7,11] or for singlet-triplet mixed superconducting phase [12]. The reason for the latter is that the upper critical magnetic fields in the  $(\text{TMTSF})_2\text{PF}_6$  strongly exceed [5-7] the Clogston-Chandrasekhar paramagnetic limit [13] and that the Knight shift in superconducting phase has not been found. Another Q1D superconductor,  $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ , has recently attracted much attention since there has been shown [14] that its upper critical field, parallel to the conducting axis, five times exceeds the Clogston-Chandrasekhar paramagnetic limit. On this basis, there has been recently suggested [14-16] that the superconductor  $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$  is a triplet one with spin part of the order parameter being insensitive to the Pauli destructive effects against superconductivity. Very recently discovered superconductors  $\text{K}_2\text{Cr}_3\text{As}_3$  and  $\text{Rb}_2\text{Cr}_3\text{As}_3$  [17-19] have added potential candidates for a triplet superconducting pairing in Q1D conductors [17,18], although in Ref.[20] the exceeding of the paramagnetic limit in the  $\text{K}_2\text{Cr}_3\text{As}_3$  is prescribed to the possible many bands effects.

Note that superconductivity in the  $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$  in Ref.[14] is observed under experimental conditions, where there is an increase of resistivity near the superconducting transition temperature. In Ref. [21], this effect was prescribed to the coexistence of superconductivity with some paramagnetically limited phase - presumably charge-density-wave (CDW). Moreover, it was shown that at very high magnetic fields,  $H \simeq 45 \text{ T}$ , where the possible CDW is destroyed by the Pauli spin-splitting

effects, some extremely low conductive state appears at  $T_c \simeq 10 \text{ K}$  [21]. In Ref.[21], it was put forward a hypothesis that it may be a triplet superconducting phase. The goal of our paper is to show that parallel to the conducting plane and conducting axis magnetic field can preserve triplet superconductivity if the coherence length, perpendicular to the plane, is less than the inter-plane distance. In this context, we pay attention that the opposite case is considered by us in Ref.[15], where the parallel magnetic field destroys superconductivity. We discuss how our current theoretical results support the hypothesis [21] about the existence of ultrahigh magnetic field triplet superconductivity as well as give a hint that this superconducting phase may survive even at higher magnetic fields in the  $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ . As a result, we suggest to perform more experiments in steady magnetic fields of the order of  $H \simeq 50 \text{ T}$  and in pulsed magnetic fields of  $H > 50 \text{ T}$ .

Let us consider a tight binding model for a Q1D layered electron spectrum:

$$\epsilon(\mathbf{p}) = -2t_x \cos(p_x a_x) - 2t_y \cos(p_y a_y) - 2t_z \cos(p_z a_z), \quad (1)$$

where  $\hbar \equiv 1$  and  $t_x \gg t_y \gg t_z$ . Note that the latter inequality allows us to linearize electron spectrum near two sheets of Q1D Fermi surface (FS), which can be expressed as:

$$p_x(p_y) = \pm p_F \pm \frac{2t_y}{v_F} \cos(p_y a_y), \quad (2)$$

where  $\pm$  stands for the right (left) sheet of the FS. Let us introduce in-plane velocity component, which is perpendicular to the conducting axis,

$$v_y(p_x) = \partial \epsilon(\mathbf{p}) / \partial p_y = 2t_y a_y \sin[p_y(p_x) a_y]. \quad (3)$$

As mentioned above the energy spectrum (1) can be linearized near the left and right sheets of the FS. Measured with respect to the Fermi energy,  $\delta \epsilon = \epsilon - \epsilon_F$ , the linearized electron spectrum (1) takes the form

$$\delta \epsilon^\pm(\mathbf{p}) = \pm v_y(p_y) [p_y - p_y^\pm(p_x)] - 2t_z \cos(p_z a_z). \quad (4)$$

In a magnetic field, parallel to the conducting  $\mathbf{x}$  axis, it is convenient to choose the corresponding vector potential in the following form:

$$\mathbf{A} = (0, 0, Hy), \quad \mathbf{H} = (H, 0, 0). \quad (5)$$

To obtain Hamiltonian in the magnetic field (5) for the electron spectrum (4), the so-called Peierls substitution method is used:

$$p_y - p_y^\pm(p_x) \rightarrow -i \frac{\partial}{\partial y} \quad \text{and} \quad p_z \rightarrow p_z - \frac{e}{c} A_z, \quad (6)$$

where  $A_z = Hy$  is the  $z$  component of the vector potential;  $e$  is the electron charge and  $c$  is the velocity of light, correspondingly. As a result, the following Hamiltonian is obtained:

$$\delta\hat{\epsilon}^{(\pm)} = \mp i v_y [p_y^\pm(p_x)] \frac{\partial}{\partial y} - 2t_z \cos\left(p_z a_z - \frac{\omega_z}{v_F} y\right), \quad \omega_z = e H a_z v_F / c, \quad (7)$$

where we do not take into account an electron spin, since below we consider only those triplet superconducting phases, which are not sensitive to the Pauli spin-splitting effects in a magnetic field. The total orbital electron wave functions, which are both eigenfunctions of energy and parallel momentum with the eigenvalues being  $\delta\epsilon$  and  $p_x$ , can be written in the factorized form

$$\Psi_{\delta\epsilon, p_x}^\pm(x, y, p_z) = e^{\pm i p_x x} e^{\pm i p_y^\pm(p_x) y} \psi_{\delta\epsilon, p_x}^\pm(y, p_z). \quad (8)$$

Here, wave functions in the mixed representation,  $\psi_{\delta\epsilon, p_x}^\pm(y, p_z)$ , are the solutions of the following Schrödinger-like equation:  $\delta\hat{\epsilon}^\pm \psi_{\delta\epsilon, p_x}^\pm(y, p_z) = \delta\epsilon \psi_{\delta\epsilon, p_x}^\pm(y, p_z)$ , which can be written as

$$\mp i v_y [p_y^\pm(p_x)] \frac{\partial \psi_{\delta\epsilon, p_x}^\pm(y, p_z)}{\partial y} = \left[ \delta\epsilon + 2t_z \cos\left(p_z a_z - \frac{\omega_z}{v_F} y\right) \right] \times \psi_{\delta\epsilon, p_x}^\pm(y, p_z). \quad (9)$$

It is important that Eq.(9) can be analytically solved:

$$\psi_{\delta\epsilon, p_x}^\pm(y, p_z) = \exp\left[\pm i \frac{2t_z}{v_y [p_y^\pm(p_x)]} \int_0^y \cos\left(p_z a_z - \frac{\omega_z}{v_F} y'\right) dy'\right] \times \exp\left(\pm i \frac{\delta\epsilon y}{v_y [p_y^\pm(p_x)]}\right). \quad (10)$$

Therefore, the total normalized wave functions (8) are:

$$\Psi_{\delta\epsilon, p_x}^\pm(x, y, p_z) = \frac{e^{\pm i p_x x} e^{\pm i p_y^\pm(p_x) y}}{\sqrt{2\pi |v_y [p_y^\pm(p_x)]|}} \exp\left(\pm i \frac{\delta\epsilon y}{v_y [p_y^\pm(p_x)]}\right) \times \exp\left[\pm i \frac{2t_z}{v_y [p_y^\pm(p_x)]} \int_0^y \cos\left(p_z a_z - \frac{\omega_z}{v_F} y'\right) dy'\right]. \quad (11)$$

Since the wave functions are known (11), we can calculate the finite temperature Green's functions by means of the standard procedure [22]:

$$G_{i\omega_n}^\pm(\mathbf{r}, \mathbf{r}') = \sum_{\delta\epsilon} \frac{[\Psi_{\delta\epsilon}^\pm(\mathbf{r}')]^* \Psi_{\delta\epsilon}^\pm(\mathbf{r})}{i\omega_n - \delta\epsilon}, \quad (12)$$

where  $\omega_n = 2\pi T(n + 1/2)$  is the so-called Matsubara's frequency.

In this paper, we consider the simplest triplet scenario of superconductivity in the  $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ , which corresponds to equal spin pairing with fully gapped Fermi surface [15]:

$$\hat{\Delta}(p_x, y) = \hat{I} \text{sgn}(p_x) \Delta(y), \quad (13)$$

where  $\hat{I}$  is a unit matrix in spin-space,  $\text{sgn}(p_x)$  takes into account the fact that the triplet superconducting order parameter (13) changes its sign on two slightly corrugated sheets of the Q1D FS,  $\Delta(y)$  is due the orbital effects against superconductivity in a magnetic field. It is important that the triplet order parameter (13) is not sensitive to the Pauli spin-splitting destructive effects in a magnetic field.

Let us make use of the Gor'kov's equations for unconventional superconductivity [4,23-25] to derive the so-called gap equation for superconducting order parameter  $\Delta(y)$  in Eq.(13). As a result of lengthly but straightforward calculations, we obtain:

$$\Delta(y) = g \left\langle \int_{|y-y_1| > \frac{|v_y(p_y)|}{\Omega}} \frac{2\pi T dy_1}{v_y(p_y) \sinh\left[\frac{2\pi T |y-y_1|}{v_y(p_y)}\right]} \Delta(y_1) \times J_0 \left\{ \frac{8t_z v_F}{\omega_z v_y(p_y)} \sin\left[\frac{\omega_z(y-y_1)}{2v_F}\right] \sin\left[\frac{\omega_z(y+y_1)}{2v_F}\right] \right\} \right\rangle_{p_y} \quad (14)$$

where  $\langle \dots \rangle_{p_y}$  stands for the averaging procedure over momentum component  $p_y$ ,  $g$  is the effective electron coupling constant, and  $\Omega$  is the cutoff energy. We pay attention that Eq.(14) is very general and was derived by us earlier in Refs. [10,15]. Indeed, Eq.(14) contains several different limiting cases for the different regions of its parameters. First of all, for low magnetic fields, it reduces to the Ginzburg-Landau equation [26] and this limiting case will be considered below as a part of more general Lawrence-Doniach-like description [27,28]. At extremely high magnetic fields, where

$$\omega_z(H) \geq \frac{8t_z v_F}{v_y^0}, \quad v_y^0 = 2t_y a_y, \quad (15)$$

or very low temperatures,

$$T \leq \tilde{T}(H) \simeq \frac{\omega_z(H) v_y^0}{2\pi^2 v_F}, \quad (16)$$

quantum effects of electron motion in a magnetic field become strong [2] and novel the so-called Reentrant Su-

perconductivity phase may appear [4,25,29]. If the inequalities (15),(16) are not fulfilled and, therefore,

$$\omega_z(H) \ll \frac{8t_z v_F}{v_y^0}, \quad v_y^0 = 2t_y a_y, \quad (17)$$

$$T \gg \tilde{T}(H) \simeq \frac{\omega_z(H) v_y^0}{2\pi^2 v_F}, \quad (18)$$

then two different physical pictures are possible: (a) anisotropic 3D superconductivity, where superconductivity disappear at high enough magnetic fields, (b) the Lawrence-Doniach-like description, where, as shown below, triplet superconductivity (13) survives at arbitrary high magnetic field. The case (a) corresponds to situation, where perpendicular to the plane coherence length,  $\xi_z$ , is bigger than the inter-plane distance,  $a_z$ , and has been already considered in Ref.[15]. The case (b), corresponding to situation, where the perpendicular coherence length is less than the inter-plane distance,

$$\xi_z^2 \ll a_z^2, \quad (19)$$

where according to Ref. [15],

$$\xi_z = \frac{\sqrt{7\zeta(3)} t_z a_z}{2\sqrt{2}\pi T_c}, \quad (20)$$

is considered below. Here,  $\zeta(x)$  is the so-called Riemann zeta-function.

It is possible to show that under conditions (17),(18) the integral Eq.(14) can be rewritten in the following way [15]:

$$\Delta(y) = g \left\langle \int_{|y-y_1| > \frac{|v_y(p_y)|}{\Omega}} \frac{2\pi T dy_1}{v_y(p_y) \sinh\left[\frac{2\pi T |y-y_1|}{v_y(p_y)}\right]} \times J_0 \left\{ \frac{4t_z(y-y_1)}{v_y(p_y)} \sin \left[ \frac{\omega_z(y+y_1)}{2v_F} \right] \right\} \Delta(y_1) \right\rangle_{p_y}. \quad (21)$$

Then, if inequality (19) is also fulfilled, it is easy to make sure that the integral (21) converges at small values of variable  $y - y_1 \sim v_y^0/(2\pi T_c)$ , where  $T_c$  is superconducting transition temperature in the absence of a magnetic field. Therefore, we can further simplify the Bessel function,

$$J_0 \left\{ \frac{4t_z(y-y_1)}{v_y(p_y)} \sin \left[ \frac{\omega_z(y+y_1)}{2v_F} \right] \right\} \simeq 1 - \frac{2t_z^2(y-y_1)^2}{v_y^2(p_y)} + \frac{2t_z^2(y-y_1)^2}{v_y^2(p_y)} \cos \left( \frac{2\omega_z y}{v_F} \right), \quad (22)$$

and expand superconducting order parameter  $\Delta(y_1)$  into a series of small parameter  $y - y_1$ :

$$\Delta(y_1) \simeq \Delta(y) + (y_1 - y) \frac{d\Delta(y)}{dy} + \frac{1}{2} (y_1 - y)^2 \frac{d^2\Delta}{dy^2}. \quad (23)$$

As a result, after some lengthly but straightforward calculations, we obtain:

$$-\xi_y^2 \frac{d^2\Delta}{dy^2} + 2 \left( \frac{\xi_z}{a_z} \right)^2 \left[ 1 - \cos \left( \frac{2\omega_z y}{v_F} \right) \right] + \left( \frac{T - T_c}{T_c} \right) \Delta(y) = 0, \quad (24)$$

where

$$\xi_y = \frac{\sqrt{7\zeta(3)} t_y a_y}{2\sqrt{2}\pi T_c}. \quad (25)$$

Note that to derive Eq.(24) from Eqs.(21)-(23), we have used the following relationships:

$$1 = g \int_{v_F/\Omega}^{\infty} \frac{2\pi T_c dx}{v_F \sinh \left( \frac{2\pi T_c x}{v_F} \right)} \quad (26)$$

and [30]

$$\int_0^{\infty} \frac{x^2 dx}{\sinh(x)} = \frac{7\zeta(3)}{2}, \quad (27)$$

It is important that, from mathematical point of view, the obtained equation (24) is similar to equations, obtained in the Lawrence-Doniach model [27,28]. Nevertheless, we pay attention that there are two main important differences between our results and the results of Refs.[27,28]. First of all, we consider Q1D superconductor in a magnetic field, parallel to its conducting axis, whereas in Refs.[27,28] only a Q2D superconductor is studied. Second, we have derived Eq.(24) from very general Eq.(14) - not from phenomenological Lawrence-Doniach model [27,28].

Note that Mathieu equation (24) is studied in details in the existing literature (see, for example, Refs.[27,28].) In the near vicinity of superconducting transition temperature,  $(T_c - T)/T_c \ll (\xi_z/a_z)^2$ , Eqs.(24) leads to the Ginzburg-Landau formula for the upper critical magnetic field,

$$H_{c2}^x(T) = \frac{\phi_0}{2\pi \xi_y \xi_z} \left( \frac{T_c - T}{T_c} \right) = \frac{4\pi^2 c T_c^2}{7\zeta(3) e t_y t_z a_y a_z} \left( \frac{T_c - T}{T_c} \right), \quad (28)$$

where  $\phi_0 = \pi c/e$  is the magnetic flux quantum. Note that the Ginzburg-Landau case corresponds to relatively low magnetic fields,

$$H \ll H^* = \frac{\phi_0 \xi_z}{2\pi a_z^2 \xi_y} \quad (29)$$

On the other hand, if magnetic field is high enough,

$$H \geq H^*, \quad (30)$$

then a magnetic field fully penetrates between the conducting layers. Mathematically, this corresponds to the case, where  $\cos(\dots)$  potential averages in the Schrödinger-like equation (24) and, therefore, in this case

$$\lim_{H/H^* \rightarrow \infty} T_c(H) \rightarrow T_c^* = T_c [1 - 2(\xi_z/a_z)^2] > 0. \quad (31)$$

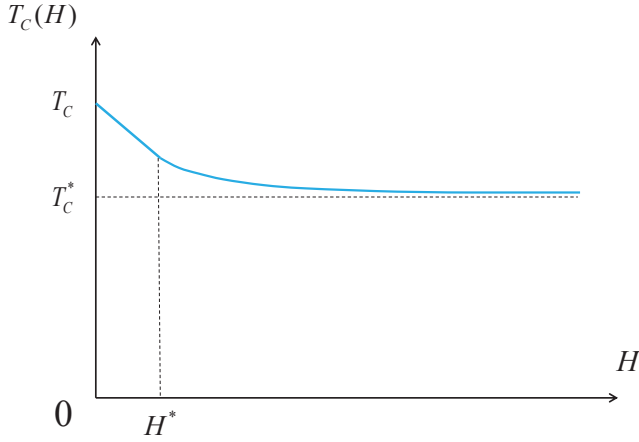


FIG. 1: Magnetic field dependence of superconducting transition temperature,  $T_c(H)$ , derived from Eq.(24), is sketched by solid line. Characteristic magnetic field,  $H^*$ , and temperature,  $T_c^*$ , are given by Eq.(29) and Eq.(31), respectively.

The overall dependence of superconducting transition temperature on a magnetic field is sketched in Fig.1, where it is clear that, under condition (19), the triplet superconducting phase (13) is stable at arbitrary high magnetic fields.

Let us demonstrate that inequalities (17)-(19), used in the paper for the derivation of Eq.(24), are likely fulfilled in the Q1D conductor  $Li_{0.9}Mo_6O_{17}$  at experimental parameters of Ref.[21], where  $H \simeq 45$  T and  $T_c \simeq 10$  K. Indeed, if we make use of the following electron band parameters, evaluated by us before in Ref.[15] (see Table I),  $t_y \simeq 41$  K and  $t_z \simeq 14$  K, and known values of the crystalline lattice parameters,  $a_z = 9.5$  Å and  $a_y = 12.7$  Å, then we obtain that Eq.(17) is valid at  $H \ll 700$  T. Moreover, using the value of  $v_y^0 = 1.4 \times 10^6$  cm/s [15], we find that  $\tilde{T} \simeq 0.3$  K. So Eqs.(17),(18) are justified very well. Let us estimate the parameter  $(\xi_z/a_z)^2$ , which has to be small for validity of Eq.(24). To this end, we use Eq.(20) and the value  $T_c \simeq 10$  K. As a result, we obtain the following values:  $(\xi_z/a_z)^2 \simeq 0.2$  and

$$T_c^* \simeq 6 \text{ K}, \quad (32)$$

which is in general agreement with the experiment [21]. It is interesting also to define how far from the Ginzburg-Landau region the possible superconductivity exists at  $H \simeq 45$  T in the  $Li_{0.9}Mo_6O_{17}$  [21]. Our estimation by means of Eq.(29) shows that the parameter  $H^*$  is equal to  $H^* \simeq 95$  T, therefore, experimental superconductivity probably corresponds to the Ginzburg-Landau region or exists close to it (see Fig.1).

To summarize, we have derived very general Eq.(14) to describe behavior of superconductivity in a layered Q1D conductor in a parallel magnetic field. We have considered in detail the particular case, where Eq.(14)

can be reduced to the Mathieu equation (24), which solution is known. We have considered the case, where perpendicular to the conducting plane coherence length,  $\xi_z$ , is much less than the inter-plane distance,  $a_z$ , in the Mathieu equation (24), and showed that, in this case, superconductivity is stable at arbitrary high magnetic fields (see Fig.1). We have calculated the corresponding parameters for the Q1D superconductor  $Li_{0.9}Mo_6O_{17}$ , using the results of Refs.[15,21], and demonstrated that our theory is very likely applicable for it at the experimental field  $H \simeq 45$  T [21]. Therefore, we conclude that very low resistive state, discovered in Ref.[21] and tentatively prescribed to superconductivity, is very likely indeed superconducting phase. We stress that very low conducting phase in Ref.[21] is observed at  $T_c \leq 10$  K and magnetic field  $H \simeq 45$  T, which is more than two times high than the the Clogston-Chandrasekhar paramagnetic limit for singlet superconductivity [13], therefore, the possible superconductivity, discovered in Ref.[21], is probably of a triplet nature. Nevertheless, at this point, we cannot completely exclude a singlet nature of the possible superconducting phase, since, in Q1D superconductors, singlet superconductivity can exist above the Clogston-Chandrasekhar paramagnetic limit in the form of Larkin-Ovchinnikov-Fulde-Ferrell phase [31-33,10,11]. More experimental works are needed to justify the nature of possible superconductivity at  $H \simeq 45$  T as well as to explore the possibility, suggested in this paper, that superconductivity can be stable at arbitrary high magnetic fields.

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